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Progress Report on
Contract DA-18-064-CML-1860

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NEW YORK UNIVERSITY
Institute of Mathematical Sciences

PROGRESS REPORT ON CONTRACT DA-18-064-CML-1860

by Ignace I. Kolodner

This report represents results obtained at the Institute of Mathematical Sciences under the auspices of the Department of the Army Chemical Corps, Contract No. DA- 18- 064- CML- 1860 .

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Introduction

This report reviews the progress of work performed under contract on "The Formation and Subsequent Behavior of Aerosols" between U. S. Army Chemical Corps and the Institute of Mathematical Sciences (formerly Institute for Mathematics and Mechanics), New York University. The period covered is November 4, 1951 - December 31, 1953.

Except for the First Quarterly Progress Report published on May 27, 1952, it has been our practice to submit at irregular intervals, reports in the form of papers discussing finished pieces of research. This has been a satisfactory practice in the early stages of this research, when only certain simple topics connected with the general subject were considered, in order to get some idea of results to be expected and to develop appropriate mathematical techniques to handle more involved parts.

The "early" stage is now practically over. During the past year we have obtained certain important results which allow us to treat the subject with some generality. With this experience we are now able to consider the more involved parts of the subject and have therefore increased the staff of the group working under the auspices of this contract. However, because of the increased mathematical difficulty of the problems now considered, publishable results will be obtained more slowly. Consequently, in addition to submitting reports in the form of papers, we shall hereafter revert to the practice of publishing Quarterly Progress Reports, briefly reviewing whatever work is in progress, even though it has not been completed. This, it is believed, will enable other groups studying Aerosols to keep in touch with progress achieved here.

Three phases can be distinguished in the Theory of Aerosols. The first is that of Aerosol Formation which is discussed in Part I. Once a suspension is formed, drops will decrease in size due to evaporation, or grow because of condensation of vapor on them. This phase is discussed in Part II. Finally, drops will collide and coalesce due to their motion produced by

turbulence or falling and also fall out of the air. We are now about to begin a study of this phase.

In the final section we list the staff participating in this research and discuss their attitudes.

Numbers in brackets refer to references. Items marked by a star (*) were published under the auspices of this contract.

Part I. The Hydrodynamics of Aerosol Formation

I-1. Review of previous research[1].

Several mechanisms which account for the breakup of bulk liquid into drops which may then form an aerosol, are more or less understood at present. The first of these -- the formation of a drop at the end of a capillary tube or eye dropper -- which depends upon gravity in addition to the surface tension and density of the liquid, and the tube diameter, operates too slowly to be of interest in the present connection. The second and most common mechanism is the breakup of a steady jet or stream which issues from some kind of hole or nozzle, as in an ordinary atomizer. The hydrodynamic theory of this type of breakup is based on the fact that the unbroken steady flow is unstable. This means that slight disturbances of the surface introduced at some point of the jet will grow in amplitude in the direction of flow until the amplitude of the surface disturbance is large enough for opposite surfaces of the jet to touch each other, thus pinching off the jet and forming drops.

A theory of this type of breakup was given by Plateau [1],

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[1] From an unpublished review on "The Hydrodynamics of Aerosol Formation" by J. B. Keller.

and greatly improved by Rayleigh [2]. The theory shows that, for a jet of circular cross-section, there is a certain range of wavelengths of disturbance for which the disturbance amplitude grows. This is called the unstable range, and in this range a wavelength equal to 4.51 times the jet diameter corresponds to the greatest growth rate. This is the wavelength of maximum instability, and it is this wavelength which accounts for the breakup and thus determines the resulting drop size. The amplitude grows according to the factor $e^{\left(\frac{T}{3\rho a^3}\right)^{1/2} \frac{z}{v}}$. Here T is the surface tension, ρ the density, a the radius and v the velocity of the jet, while z is distance along the jet. This formula accounts for the observations of Savart [3] on the length of the continuous, unbroken, portion of a jet. It is also possible to apply this theory to determine the periodic changes in shape of a slightly non-circular jet, originally studied by Bidone, which were accounted for in a somewhat cruder manner by Magnus [4] and Buff [5] . The theory of circular jet breakup has also been extended to jets of viscous liquid and to jets of air in liquid by Rayleigh [6],[7], and to charged jets by Basset [8].

Rayleigh [9] has also given a two-dimensional theory of the instability of a plane interface between two fluids in relative motion, the fluids being either bounded or extending to infinity in the direction perpendicular to the interface. He also considered the instability of a two-dimensional jet, and examined the effect of varying the profile of the steady flow.

A third mechanism of drop formation acts when an explosive surrounded by a liquid layer, is detonated. As Taylor [10]has shown, liquid surfaces may be unstable when accelerated normal to themselves, and it is this instability which operates in explosives or some other transient devices. Taylor's theory of this instability has been experimentally verified by Lewis [11].

After drops have been formed, they may possess enough energy of vibration or deformation to break into still smaller drops. A start on the theory of this phenomenon was made by

Rayleigh [12], and also by Webb [13], who determined the small vibrations of spherical drops. It was further pursued by Rayleigh [14] who took account of the presence of charge on the drop and showed that enough charge made the drop unstable.

If drops are subjected to a steady air flow or to a shock wave, they may also break up. In England the Porton experiments demonstrate these effects, and rough theories have been given by Penney and by Taylor in Porton reports.

I-2 Breakup of steady jets.

On the basis of Rayleigh's work, it seems that the breakup of most steady streams from nozzles is due to instability governed by surface tension and inertia. It should be possible to analyze the breakup of various fluid sheets, such as the "tulip" formed by a conical nozzle, by appropriate modification of Rayleigh's theory. In order to do this, it is necessary to obtain first an exact or approximate solution of the steady, undisturbed flow. Since the exact solution of jet problems is difficult, various approximate solutions, such as that given by the hydraulic theory, will have to be used.

In line with this program Keller and Weitz [15*] developed a systematic method for handling thin heavy jets, both steady and unsteady. They obtain formal expansions in powers of a thickness parameter, the zero order terms being identical with the hydraulic approximation. The method is similar to that used by Friedrichs [16], and Keller [17] in the shallow water theory, and although it is not known whether these expansions converge, the recent proof of existence of a solitary wave in channels, based on a similar expansion[2], indicate that they may converge.

The next step would be to discuss the time dependent pertur-

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[2] By Friedrichs, K. O. and D. Hyers; to appear in the Communications on Pure and Applied Mathematics.

bations on the zero order solutions of [15*] in a manner similar to that discussed in I-3. We plan to carry this out for the flow issuing from a vertical nozzle, and for the "tulip" flow formed by a conical nozzle. This research will be continued after the completion of the problems outlined in I-3 now under consideration. See also the discussion in I-4.

I-3 Study of breakup of thin liquid layers.

The ideas of Taylor [10] have been applied by Keller and Kolodner [18*] in studying drop formation. They consider a plane liquid layer, accelerated in a direction perpendicular to the layer surface, by imposing a pressure difference on opposite surfaces. (The origin of this pressure difference need not be specified) The zero order solution is a parallel flow, with velocity of any of the two bounding surfaces, which is also an exact solution. It is then argued that the flow deviates from the zero order solution because either the bounding surfaces are not perfectly plane or the pressure on the boundary is not exactly constant. A first order perturbation is then considered and it is shown that to this order the surface may be imagined to be divided into rectangles by two sets of waves in perpendicular directions. Alternate rectangles are raised and lowered, and for certain dimensions the amplitude grows until opposite faces of the liquid layer touch, leading to breakup into drops the size of which depends upon rectangle size. A certain set of rectangles leads to maximum instability, and determines the drop size, just as in the theory of jets. The formula obtained for the drop radius r is

$$r = \left(\frac{9\pi T h^2}{2 |p_2 - p_1|} \right)^{1/3}$$

where,

h = thickness of the layer
T = coefficient of surface tension
$p_2 - p_1$ = difference of pressures executed on opposite sides.

The time of breakdown is also estimated.

As is seen from the formula, the assumption that there is surface tension is essential in this theory. The theory was constructed under the assumption that the liquid is incompressible and inviscid. We also attempted to consider the effects of viscosity, but this led to complications which we were unable to resolve. This attempt was consequently shelved, but will be reconsidered at a later date.

There is doubt as to the range of applicability of the formula quoted above, and a more refined approach to problems of this type is now under investigation (see II-4). Nevertheless, the simplicity of the method used in [18*] warrants its application to the problem of accelerated spherical and cylindrical layers.

The spherical case is now near completion, and barring unforseen difficulties, a report on this piece of research will be published within a month[3]. A hint on the method of solution was supplied in [19*], section III[4]. The problem possesses a great deal of similarity to that of underwater explosion bubbles, and some of the techniques developed for treatment of the latter by Keller and Kolodner [20] are used. The study of cylindrical layer will follow that of spherical layer.

I-4 Rigorous treatment of motion of liquid layer

We suggested that the formula of Part I-3, inasmuch as it is of considerable practical interest, be tested by exploding into liquid sheets, and thus accelerating them into air. To obtain a variety of drop sizes, we suggested that the lower surface of the layer be initially deformed in a controlled way, for example, by introducing slight dimples distributed periodically.

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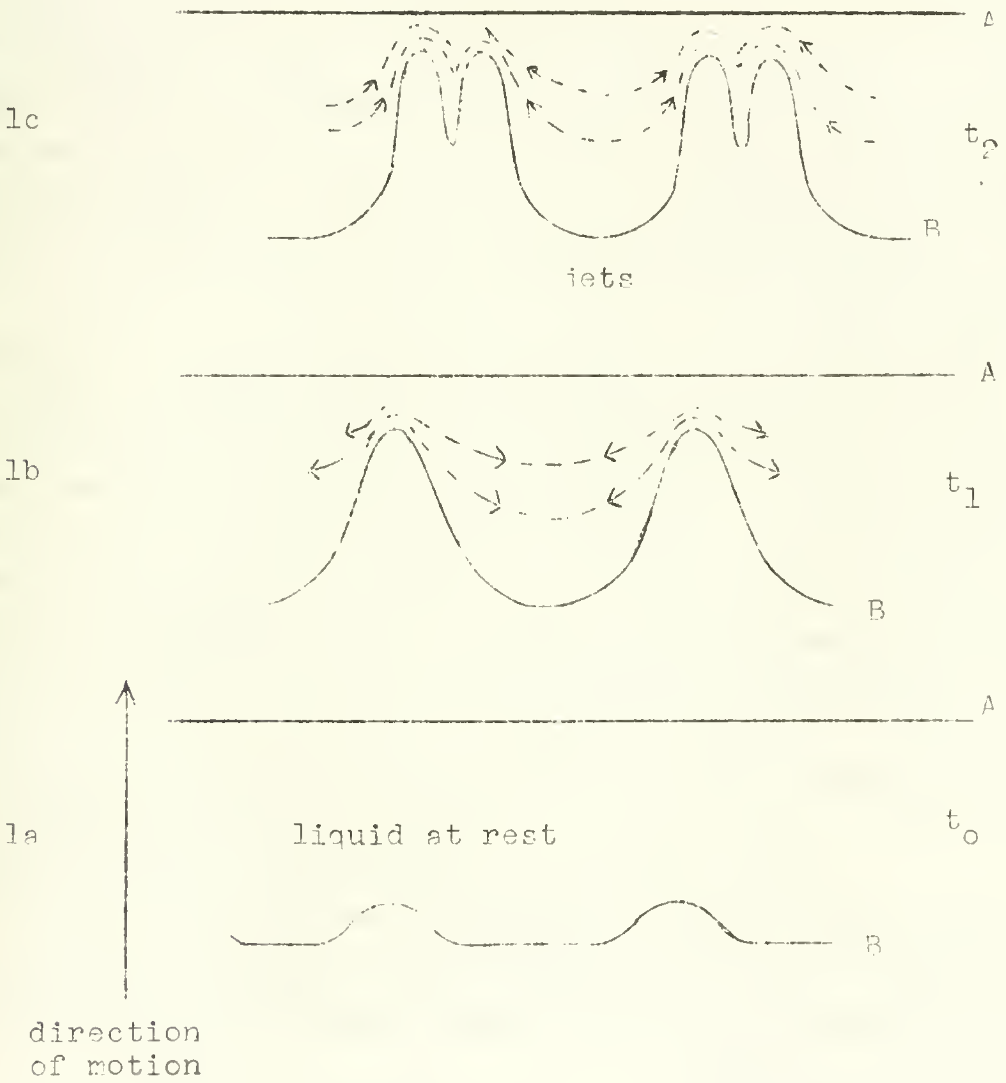
[3] by Keller, J. B., I. I. Kolodner and M. Milgram.

[4] With some errors. At that time we were unaware of the importance of surface tension in problems of this type.

Such experiments were carried out by Holland[5] at Camp Dietrich, Maryland, and in the cases tested the initial dimples deepened at first, whereas at a later time small jets would emanate from the troughs of the dimples in the direction opposite to that of motion of the layer. See Fig. 1 a, b, c.

Figure 1



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[5] Oval communication by Dr. T. Holland.

In our opinion, these results do not necessarily reflect on the validity of the formula of section I-3, nor do they indicate that the mechanisms proposed by Rayleigh and Taylor to account for the breakup of bulk liquid cannot lead to results in agreement with experiment. Our treatment applies to thin jets and layers, but the mathematical techniques used are too crude to account for the finer features observed by Holland, or to predict the maximum thickness of layer for which our theory applies. It has been suggested that compressibility effects are responsible for the appearance of jets in the case tested. This, in our opinion, is not necessarily so, for a jet will form whenever two streams impinge on each other and this seems to be the situation in Fig. 1c.

In any case, since our theory does not explain Holland's experiments, a better theory is called for. The discrepancies are due either to wrong physical assumptions about the liquid - such as incompressibility, or to too crude mathematical treatment - extrapolating the method of perturbations to account for large scale phenomena used in [18*]. Accounting for compressibility effects will result in mathematical complications which cannot be mastered at present, and , in view of the argument of the previous paragraph, should not lead to an essential improvement of the theory. On the other hand, a vigorous treatment of motion of accelerated layers should lead to a clear-cut answer to whether the incompressibility assumption is satisfactory, and if so, should shed more light on how the breakup occurs.

This problem is very complicated and for simplicity we consider as a test problem a two dimensional liquid layer extending to infinity, with a single dimple on the lower surface accelerated upwards; see Fig. 2. Placing the y direction in the direction of acceleration, the equation of the upper surface is

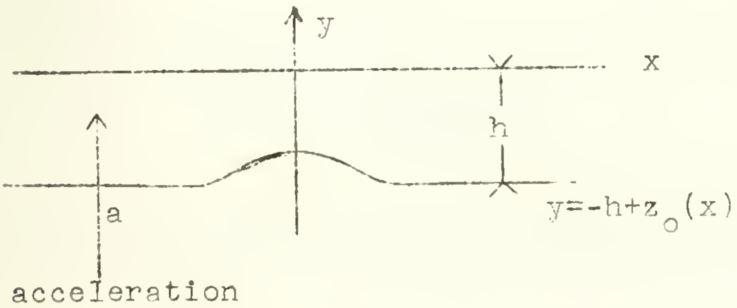

Figure 2

$$y = \frac{1}{2} at^2 \quad ,$$

and that of the lower surface is initially

$$y = - h + z_o(x) \quad .$$

We assume that z_o is an even function of x, and that for $x > 0$ it decreases monotonically from some positive value $\sigma = z_o(0)$, $\sigma < h$, to zero. The problem is to find the boundary function $z(x,t)$(in which we are primarily interested), and the velocity potential $\phi(x,y,t)$, which satisfy the following equations:

$$(1) \quad \Delta\phi = 0 \ , \text{ for } \frac{1}{2}at^2 - h + z(x,t) < y < \frac{1}{2} at^2$$

$$(2) \quad \phi_y = at \quad \text{for} \quad y = \frac{1}{2} at^2$$

$$\left.\begin{aligned} &(3) \quad \phi_y - z_x\phi_x = at + z_t \\ &(4) \quad \rho(\phi_t + \tfrac{1}{2}(\nabla \phi)^2) + p_o - 2TH = f(t) \end{aligned}\right\} \text{for } y = \tfrac{1}{2}at^2 - h + z(x,t)$$

$$(5) \quad |\nabla \phi| \longrightarrow 0 \quad \text{as } |x| \longrightarrow \infty$$

$$(6) \quad \begin{aligned} z(x,0) &= z_o(x) \\ z_t(x,0) &= 0 \quad . \end{aligned}$$

Here ρ is the density of liquid, p_o the pressure of adjacent air, T the coefficient of tension, and H the mean curvature of the upper surface;

$$H = - \frac{z_{xx}}{2(1+z_x^2)^{3/2}} \quad .$$

The function f(t) is an arbitrary function of time. Introducing new variables $\bar{y}$ and $\Phi(x,\bar{y},t)$ defined by

$$y = \frac{1}{2} at^2 + \overline{y}$$

$$\phi = at\overline{y} + \overline{\Phi}(x,\overline{y},t)$$

the first four equations become on dropping bars,

$$(1') \quad \Delta\phi = 0 \quad \text{for} \quad -h + z(x,t) < y < 0$$

$$(2') \quad \phi_y = 0 \quad \text{for} \quad y = 0$$

$$\left.\begin{array}{ll} (3') & \phi_y - z_x\phi_x = z_t \\ (4') & \phi_t + \frac{1}{2}(\nabla\phi)^2 + az + \frac{T}{\rho}\frac{z_{xx}}{(1+z_x^2)^{3/2}} = 0 \end{array}\right\} \text{for } y = -h+z(x,t)$$

whereas the last two remain unchanged. Here we set

$$f(t) = p_o - \frac{1}{2} g^2t^2 + \rho h \quad ,$$

for convenience.

The problem thus formulated is a free boundary value problem for the potential equation to which the recently developed method for handling such problems[6] may be applied. This method consists in reduction of the problem to a system of two integro-differential equations for the unknown boundary function $z(x,t)$ and for the potential on the boundary $\psi(x,t) = \phi(x,-h+z(x,t),t)$. In our case these equations are:

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[6] By Kolodner, I. I., to appear in Communications on Pure and Applied Mathematics.

$$(7)\begin{cases} \psi_t + \dfrac{\psi_x^2 - 2\psi_x z_x z_t - z_t^2}{2(1+z_x^2)} + az + \dfrac{T}{\rho}\, \dfrac{z_{xx}}{(1+z_x^2)^{3/2}} = 0 \\ \psi(x,t) = \dfrac{1}{\pi} \displaystyle\int_{-\infty}^{\infty} \psi(\xi,t) \dfrac{\partial}{\partial\xi}\left(\tan^{-1} \dfrac{z(\xi,t)-z(x,t)}{\xi - x} + \tan^{-1} \dfrac{z(\xi,t)+z(x,t)-2h}{\xi - x}\right) d\xi \\ + \dfrac{1}{\pi} \displaystyle\int_{-\infty}^{\infty} z_t(\xi,t) \Big\{ \log[(\xi-x)^2 + (z(\xi,t)-z(x,t))^2][(\xi-x)^2 + (z(\xi,t) + \\ \qquad\qquad + z(x,t)-2h)^2] \Big\}\, d\xi \end{cases}$$

subject to the initial conditions (6) .

These equations are now under study, but because of their complexity this may take a considerable amount of time. We expect to devise on the basis of this study an approximation procedure which is more refined than that used in [18*], and which will bring out the influence of the thickness of the layer on the type of behaviour that may occur. If this leads to a theoretical prediction in agreement with experimental results, we shall have here a basis for predicting the apperrance of small jets in addition to drops. These in turn, could be handled by the Rayleigh theory (see section I-2), and this would be a theory of formation of satellite drops. Since the three-dimensional case of a layer with several circular dimples may be handled in a similar manner (although the corresponding equations are more complicated), it is hoped that the experience thus gained in considering this test problem will be applicable to the theoretical study of experiments by Holland.

Part II. Decay of drops by evaporation and growth by condensation

II-1. Review of previous research; formulation of problems

Once the breakup of sheets or sprays is complete, drops will still decrease in size due to evaporation, or grow because of condensation of vapor on them. Thecondensation phenomenon may also be used to produce drops, which first form on dust, ions or other nuclei and then grow according to the theory to be discussed below. This is the principle of the Chemical Corps' oil smoke generator, which is also used in artificial production of rain.

Experimental investigations and approximate theoretical treatments of these effects have been described by a number of writers. For example, Langmuir [21] and Fuchs [22] treated the theory of the isolated stationary droplet, while Topley and Whytlaw-Gray [23] considered this problem experimentally. The evaporation of a moving droplet was studied experimentally by Takahasi [24] and Frössling [25], and theoretical and experimental results were given by Kinzer and Gunn [26]. The problem of the growth of a collection of aerosols or hydrosols has been treated extensively by La Mer and his associates (see e.g. [27] and [28])and by Collins and his associates (see e.g. [29] and [30]).

However, most of the theoretical work on this subject has been carried out by physical chemists who were forced to make more or less restrictive physical assumptions in order to reduce the problem to one which could be handled by standard mathematical techniques. For example, the mathematical difficulty occasioned by the moving boundary at the surface of the droplet has invariably been avoided by the introduction of additional physical restrictions. One of the main objectives of our work has been the exact mathematical treatment of the moving boundary problem, as it stands.

Our considerations are based on the following physical model. If a liquid drop is surrounded by a gas in which the pressure of the drop's vapor is below the equilibrium vapor pressure, the vapor diffuses away from the drop surface and the drop evaporates. This evaporation, which maintains the vapor pressure near the surface at the equilibrium value, results in decay of the drop. Conversely, if the vapor pressure in the surrounding gas exceeds the equilibrium value, vapor diffuses toward the drop, condenses on its surface and the drop grows. (The same description applies to a drop dissolving or growing in a liquid, if the term "vapor pressure" is replaced by "concentration of solute".) In general, the equilibrium pressure depends on temperature and on the radius of the drop. In this model, the temperature is assumed constant, and the dependence of equilibrium pressure on the drop's radius is neglected. Hence the equilibrium pressure is constant, and we take it to be the equilibrium pressure at a given temperature.

A more accurate theory of evaporation (condensation) accounting for thermal phenomena accompanying this process is discussed in section II-7.

We furthermore assume that the drop or drops remain spherical at all times due to some constraint (like surface tension) the nature of which is immaterial for the present theory. Let a_n be the initial radius of drop n centered at P_n, n = 1, 2, 3 ..., see Fig. 3. Let

g - equilibrium mass concentration
c_o - initial mass concentration of vapor
D - coefficient of diffusion of vapor into air
ρ - density of the condensed vapor.

The mathematical problem is to find the drop radii $\rho_i(t)$, and the mass concentration c(x,y,z,t) satis-

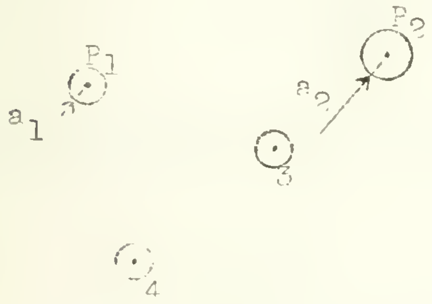

Figure 3

fying the following equations:

$$\left.\begin{array}{ll}(1) & D\Delta c = c_t \\ (2) & c(x,y,z,0) = c_o\end{array}\right\} \text{ in the vapor region}$$

$$\left.\begin{array}{ll}(3) & c = g \quad \text{on } S_n \;, \\ (4) & 4\pi(\rho-g)\dot{\rho}_n(t) = D \oiint \frac{\partial c}{\partial r_n}\, d\omega \;, \\ (5) & \rho_n(0) = a_n\end{array}\right\} \text{ for all } n.$$

Here, S_n is the surface of the n'th drop, $\frac{\partial c}{\partial r_n}$ is the derivative of concentration in direction emanating from the center of the n'th drop and evaluated on S_n, and $d\omega$ is the solid angle element. Condition (4) states that the mass of vapor flowing into the variable vapor region is equal to the mass of liquid that evaporates.

Introducing dimensionless variables defined by

$$c(x,y,z,t) = c_o + (g-c_o)\bar{c}(\bar{x},\bar{y},\bar{z},\bar{t})$$

$$x = a\bar{x}, \quad y = a\bar{y}, \quad z = a\bar{z}$$

$$t = \frac{a^2}{D}\,\bar{t}$$

$$\rho_n(t) = a\bar{\rho}_n(\bar{t}) \;, \quad a_n = a\bar{a}_n$$

$$\alpha = \frac{g-c_o}{\rho-g} \;,$$

the equations become, on dropping bars,

$$\left.\begin{array}{ll}(1') & \Delta c = c_t \\ (2') & c(x,y,z,0) = 0\end{array}\right\} \text{ in the vapor region}$$

$$\left.\begin{array}{lll} (3') & c = 1 & \text{on } S_n \\ (4') & \dot{\rho}_n = \frac{\alpha}{4\pi} \oint\!\!\!\oint \frac{\partial c}{\partial r_n}\, d\omega & \\ (5') & \rho_n(0) = a_n & \end{array}\right\} \quad \text{for all } n .$$

If $g > c_o$, the drops evaporate and $\alpha > 0$. If $g < c_o$, drops grow and $\alpha < 0$. In all practical cases, $g \ll \rho$, $c_o \ll \rho$, so that α is a small number ($\alpha = 2 \times 10^{-5}$ for water drops evaporating into dry air at 73° F). In any case, $c_o < \rho$, so that $\alpha > -1$.

In case of a single drop, c becomes a function of distance from the center of the drop, r. Choosing for the scaling factor "a" the initial radius of the drop, the equations simplify to

$$(1'') \quad \Delta c \equiv \frac{1}{r}(rc)_{rr} = c_t \,, \qquad r > \rho(t)$$

$$(2'') \quad c(r,0) = 0, \qquad r > 1$$

$$(3'') \quad c(\rho(t), t) = 1 \,, \qquad t \geq 0$$

$$(4'') \quad \dot{\rho}(t) = \alpha c_r(\rho(t), t) \,, \qquad t > 0$$

$$(5'') \quad \rho(0) = 1$$

It is worthwhile to consider also the case of one-dimensional "drop", i.e. that of evaporating slab of water. In this case, the equations become

$$(1''') \quad \Delta c \equiv c_{xx} = c_t \,, \qquad x > \rho(t)$$

$$(2''') \quad c(x,0) = 0 \,, \qquad x > 1$$

$$(3''') \quad c(\rho(t), t) = 1 \,, \qquad t \geq 0$$

$$(4''') \quad \dot{\rho}(t) = \alpha c_x(\rho(t), t) \,, \qquad t > 0$$

$(5''')\quad \rho(0) = 1$

with the additional condition, $c(x, t) = c(-x, t)$.

We shall refer to these problems as problems A, B, and C.

The three problems formulated above (in decreasing order of difficulty) are free boundary value problems for the heat equation. Problems of similar nature arise in the study of change of phase, and were first considered by Stefan [31], who solved a very special case explicitly. Stefan-like problems were considered later by Datsev [32], Evans, Isaacson and MacDonald [33], and Evans [34], but their methods and techniques turned out to be of little value in the study of problems arising in cases considered here.

II-2 Approximate treatment of the problem of a single drop.

The study of behavior of a single drop is preliminary to that of collection of drops. The single drop problem is in itself of great interest. Experimental data concerning evaporation are available and these indicate that, except for a short initial period and very small drops, the square of the drop's radius decreases linearly with time; see [24], [25], [26], Godsave [35] and Ranz and Marshall [36].

The initial study[7] was reported in [19*], section II. The one-dimensional case, problem C, can be solved explicitly and the result is

$$(1)\quad \rho(t) = 1 - 2z\sqrt{t}$$

where z is the solution of the transcendental equation

$$z = 2\alpha e^{-z^2} \int_{-z}^{\infty} e^{-\sigma^2}\, d\sigma \quad .$$

This equation has one solution provided that $\alpha > -1$ (physically, the

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[7] by Keller, J. B.

case $\alpha = -1$ corresponds to the critical point when the change of phase is not well defined). If α is small, $z \sim \frac{\alpha}{\sqrt{\pi}}$, and in case of evaporation, $\alpha > 0$, this yields for the time of evaporation t_o,

$$(1') \quad t_o \sim \frac{\pi}{4\alpha^2}$$

Although equation (1) is exact, approximation methods were also devised with intent of generalization to the three-dimensional case, problem B, where an explicit solution cannot be constructed. The first of these consists in assuming an approximation to the evaporation curve, and foregoing the second free boundary condition, eq. (1-4'''). One gets thus an ordinary boundary value problem to solve and one uses this solution in eq. (1-4''') to get an improved approximation for the evaporation curve. If one takes for the first approximation to ρ

$$\rho = \text{constant} = 1$$

(which corresponds to $\alpha = 0$), the second approximation yields

$$\rho = 1 - \frac{2\alpha}{\sqrt{\pi}} \sqrt{t} \quad ,$$

hence a result identical with (1) for small α. Iteration of this process leads to rather complicated expressions.

The second method is the so-called quasi-static approximation. One assumes that the concentration is slowly time dependent so that the term c_t in the differential equation can be at first omitted. One thus constructs a first approximation to the concentration and the evaporation curve. The exact result in the one-dimensional case shows however, that c_t is infinite for $t = 0$ at $x = 1$. Thus this method is not expected to be satisfactory for small t. Indeed in this case, it cannot be applied at all, for it leads to an expression for c which does not remain bounded as x increases to infinity.

However, both methods can be applied in the three-dimensional case, problem B. The first yields

$$(2) \quad \rho(t) = 1 - \frac{2a}{\sqrt{\pi}}\sqrt{t} - at, \qquad (2') \quad t_o \sim \frac{1}{a} - \frac{1}{\sqrt{\pi}}\frac{1}{\sqrt{a}} + \frac{2}{\pi} + \dots,$$

for a small

whereas the second gives

$$(3) \quad \rho^2(t) = 1 - 2at \quad , \qquad (3') \quad t_o \sim \frac{1}{2a}$$

The two methods thus lead to different results and predict evaporation times differing by a factor of 2 for small a. Both results are therefore inconclusive, although the first one agrees with the exact result in the one-dimensional case, whereas the second is in agreement with the experimental results in three-dimensional case.

A third approximation method was devised by Keller, Kolodner, and Ritger [37*]. They represent the concentration by an integral, the form of which is suggested by the exact solution of the one-dimensional problem. This integral satisfies the heat equation no matter what its kernel is, provided that it has the required form. Next the kernel "f" is expanded in formal powers of the variable on which it depends,

$$f(\tau) = A_o(1 + \sum_{n=1}^{\infty} A_n \tau^n) ,$$

whereas the unknown radius is assumed to have an expansion of the form

$$\rho(t) = 1 + 2\sum_{n=1}^{\infty} a_n t^{n/2} ,$$

and the coefficients are determined by satisfying the two free boundary conditions. As is shown, the two expansions can have at

best an asymptotic character near $t=0$ whereas an expression for $\rho(t)$ valid during the full evaporation period is desired. To get this, one defines as the N'th approximation to $f(\tau)$, the polynomial obtained from the series on dropping all terms from the N+1'st on, with coefficients $A_n (n=0,1,....)$ previously determined, and one defines the N'th approximation to ρ, by requiring that it satisfies the second boundary condition (1-4") with c replaced by its N'th approximation. At each step one thus gets an ordinary differential equation which must be integrated numerically. A calculation was carried out for $\alpha=2 \times 10^{-5}$, N=1,2,3, showing that the second approximation for ρ agrees with the third to 3 decimals. For α small these equations can be simplified without introducing an appreciable error, and one gets for the second approximation,

$$(4) \qquad \rho^2 \sim 1 - \frac{4\alpha}{\sqrt{\pi}}\sqrt{t} - 2\alpha t \quad ,$$

also agreeing to within 3 decimals with the computed result. The evaporation time is

$$(4') \qquad t_0 \sim \frac{1}{2\alpha} - \frac{4}{3\sqrt{2\pi}}\,\frac{1}{\sqrt{\alpha}} + O(1) \quad .$$

Formula (4) agrees with experimental results over the full range of evaporation time.

II-3 Rigorous treatment of an evaporating drop.

Of the three methods discussed in the preceding section, the third cannot be easily adopted to handle more complicated problems, whereas the first two can. Since the third method led to a result in agreement with that of the first method for small t, and with that of the second for large t, its justification would at the same time establish the respective ranges of applicability of the first two methods. With this in mind, we attacked the problem B rigorously hoping to reestablish the formula (2-4) .

Since equation (2-1") leads to a very simple ordinary differen-

tial equation for the Laplace Transform of $u(r,t) = rc(r,t)$ with respect to t, use of the Laplace Transformation seemed to be proper for a problem of this type. Evans, Isaacson and McDonald [33], expressed the hope that this technique should produce an elegant solution to their problem, but got nowhere. Likewise, in case of our problem the use of Laplace Transform led to a formal expression for the concentration function, which would be the correct expression *if the boundary ρ were known*, but could not be easily used to determine their boundary. (It was realized much later that this expression could be obtained in a simpler way by applying Green's theorem to the function u, see [38].) What evolved, however, from an interpretation of this formal result, was a basically simple, but very general and powerful method for solving the free boundary value problems for the heat equation, by Kolodner [39][8]. The method consists in a reduction of the problem to an integro-differential equation for the boundary.[9] This method was applied to the case of evaporation of a drop by Kolodner [40][10] and yields $\rho(t)$ as the solution of

$$(1)\quad \begin{cases} \sqrt{\pi}\,\rho\dot{\rho} = -2a \displaystyle\int_{-\infty}^{z(\rho,0)} \left(1 - \frac{\tau}{\sqrt{t}}\right) e^{-\tau^2}\,dt + \int_0^t \frac{\rho(\sigma)\dot{\rho}(\tau)}{t-\sigma}\, z(\rho,\sigma)\, e^{-z^2(\rho,\sigma)}\,d\sigma = G[\rho] \\ \\ \rho(0) = 1 \end{cases}$$

where

$$z(x,\sigma) = \frac{x-\rho(\sigma)}{2\sqrt{t-\sigma}}, \qquad z(x,0) = \frac{x-1}{2\sqrt{t}} \quad .$$

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[8] The reference is to an abstract of a paper presented at the April 1953 meeting of the American Mathematical Society in New York City. Details will appear in Communications on Pure and Applied Mathematics.

[9] The fundamental idea is the same as the one used by the author for free boundary value problems for the potential equation, mentioned in section I-4, which actually was developed later.

[10] An extract from a longer and rather involved paper to appear in Communications on Pure and Applied Mathematics.

[illegible]

[illegible]

[illegible]

[illegible]

[illegible]

[illegible]

This equation shows that

$$(2) \qquad \rho^2(t) \geq 1 - \frac{4\alpha}{\sqrt{\pi}} \sqrt{t} - 2\alpha \quad ,$$

whereas an independent argument yields directly from the formulation of the problem, an upper bound for ρ

$$(3) \qquad \rho^2 \leq 1 - \frac{2\alpha}{1+\alpha} t \quad ,$$

see Fig. 4. From these one gets a two sided inequality for the evaporation time t_o ,

$$(4) \quad \frac{1}{2\alpha} + \frac{2}{\pi}\left(1 - \sqrt{1 + \frac{\pi}{2\alpha}}\right) \leq t_o \leq \frac{1}{2\alpha} + \frac{1}{2}$$

For $\alpha = 2 \times 10^{-5}$, the case considered previously, either of the bounds (2) and (3) yield ρ^2 with an absolute error not exceeding .0072 over the full evaporation time, whereas (4) determines t_o with a relative error of at most .0072.

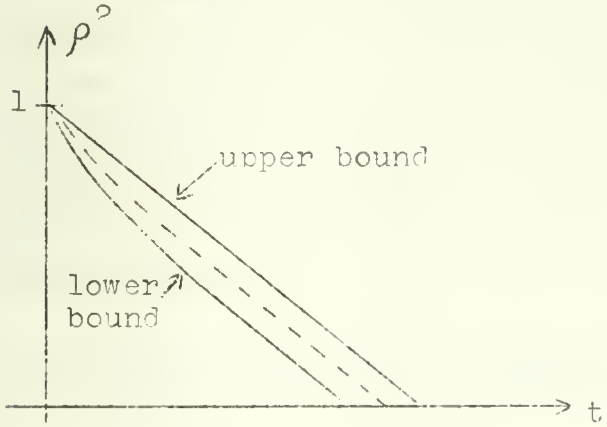

Figure 4

The lower bound for ρ^2 is identical with formula (2-4). Thus the results previously obtained are confirmed, provided that the solution of eq. (1) exists. The existence of a unique solution in *the small* can be demonstrated using standard techniques, provided that $0 \leq \alpha \leq .57$; since, however, we are interested in results for the full evaporation period, such a proof is not satisfactory. An existence and uniqueness theorem in *the large* was constructed by showing that the solution of eq. (1) is the limit of an "alternating" sequence of functions $\rho_n(t)$ defined over the interval $0 \leq t \leq t_n$ increasing with n and tending to t_o as $n \longrightarrow \infty$. Any two consecutive terms of the sequence form an upper and lower bound for the solution in the interval over which these terms are defined. Thus there is a

possibility of improving the bounds (2) and (3) which is useful when either α is larger (but smaller than .38), or when higher accuracy is desired. ρ_0 is identical with the right hand side of inequality (2), whereas ρ_1 is an upper bound which is better than eq. (3) for small t, but not as good as (3) for large t.

II-4 Growth of drop by condensation

The case of condensation differs from the case of evaporation in that $-1 < \alpha < 0$, (see section II-1). The general method discussed in [39] leads to the same equation for $\rho(t)$ as equation (3-1) with $\alpha < 0$. However, as is indicated in [39], the condition that $\rho(t)$ satisfies this equation is a necessary condition for the solvability of the free boundary value problem in case of condensation, whereas the sufficiency has not been proved. Nevertheless, it is possible to derive a second equation for $\rho(t)$, the solvability of which is fully equivalent to the solvability of the problem as posed originally. This equation is

$$(1)\quad \sqrt{\pi}\,\rho(t) = 2\rho(t)\int_{z(\rho,0)}^{0} e^{-\tau^2}d\tau - 2\sqrt{t}\,e^{-z^2(\rho,0)} - \frac{1}{\alpha}\int_0^t \frac{\rho(\sigma)\dot{\rho}(\sigma)}{\sqrt{t-\sigma}}e^{-z^2(\rho,\sigma)}d\sigma.$$

The techniques used in solving equation (3-1) do not apply in this case. and, so far, this equation has not been solved in the large. Yet, it can be shown quite independently, that

$$(2)\qquad \rho^2 \geq 1 - \frac{2\alpha}{1+\alpha}\,t, \quad -1 < \alpha < 0 \quad .$$

Probably for t large, $\rho \sim \sqrt{-\frac{2\alpha}{1+\alpha}\,t}$.

We are now, in conjunction with the study of other problems discussed in section II-5, in the process of devising new techniques, which, it is expected, will enable us to solve equation (1), and in particular will establish an upper bound in the large for ρ .

II-5 Approximate treatment of collection of drops

An approximate treatment of collection of evaporating drops based on the quasi-static approach was attempted by Keller[11]. Difficulties were encountered which could not be overcome without introducing certain additional assumptions concerning the model. As it appears now, on the basis of a more accurate treatment, (as yet incomplete), discussed in section II-6, these assumptions are unnecessary, but the method must be modified. This work will be taken up again in the future.

Another approximate approach for a collection of initially identical drops with centers at vertices of a cubic lattice stems from the consideration of a one-dimensional collection. In this case, Fig. 5, the evaporation curves are all the same, and the concentration c is a periodic function of x with period 2M = distance between two successive drops, and has the lines x = M, x = 3M, ..., x = (2n+1)M... for lines of symmetry. Clearly, on these lines, $c_x = 0$. To solve the problem, it is thus sufficient to consider a half period interval. c and ρ then satisfy equations (1-1"') through (1-5"'), except that x is restricted to the range $\rho(t) \leq x \leq M$, and for x = M, $c_x = 0$. This condition also states that the flux of vapor through the line of symmetry is 0, and therefore the problem of a one-dimensional collection is fully equivalent to that of a single drop placed in an impermeable box, with walls at distance M from the center of the drop.

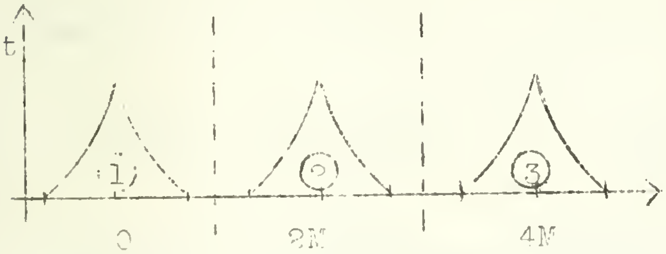

Figure 5

In the three-dimensional case the situation is similar. However, to be able to apply the general method of [39], we assume for simplicity that the box is not cubic, but spherical. This assumption, it is believed, ought to have little effect on the result.

The method of [39] leads to the following equations for the radii :

$$(1)\ \begin{cases} \sqrt{\pi}\,\dot{\rho}(t) = -\frac{a}{\sqrt{t}}e^{-z^2(\rho,0)} + \int_0^t \frac{\dot{\rho}(\sigma)}{t-\sigma} z(\rho,\sigma)e^{-z^2(\rho,\sigma)}d\sigma \\ \qquad + \frac{a}{\sqrt{t}}\,e^{-z^2(2M-\rho,0)} - \int_0^t \frac{\dot{\rho}(\sigma)}{t-\sigma} z(2M-\rho,\sigma)e^{-z^2(2M-\rho,\sigma)}d\sigma \\ \rho(0) = 1 \end{cases}$$

in the one-dimensional case, and

$$(2)\ \begin{cases} \sqrt{\pi}\,\rho(t)\dot{\rho}(t) = G[\rho] + 2a\int_0^t K(\sigma)h_x(\rho(t),t,\sigma)d\sigma \\ \rho(0) = 1 \end{cases}$$

where, $G[\rho]$ is defined in equation (3-1),

$$K(t) = \left(\sqrt{t} - \frac{M}{2\sqrt{t}}\right)e^{-z^2(M,0)} + \frac{1}{2a}\int_0^t \frac{\rho(\sigma)\dot{\rho}(\sigma)}{t-\sigma}\left(1+\frac{Mz(M,\sigma)}{\sqrt{t-\sigma}}\right)e^{-z^2(M,\sigma)}d\sigma$$

$$h(x,t,\sigma) = -\frac{1}{M\sqrt{\pi}}\left[\frac{1}{\sqrt{t-\sigma}}\exp\left(-\left(\frac{x-M}{2\sqrt{t-\sigma}}\right)^2\right) + \frac{2}{M}\exp\left(\frac{x-M}{M} + \frac{t-\sigma}{M^2}\right)\int_{-\infty}^{\frac{x-M}{2\sqrt{t-\sigma}}+\frac{\sqrt{t-\sigma}}{M}} e^{-\tau^2}d\tau\right]$$

$$z(\rho,\sigma) = z(\rho(t),\sigma),$$

and $z(x,\sigma)$ is defined in section II-3. These equations apply to the case of evaporation only, but can be used in case of condensation in the sense explained in section II-4.

These equations, as well as eq. (4-1), and some other equations (such as equations (6-4)) stemming from related problems, are now being studied, and our aim is to produce, in each case, a solution in the large by means of a single iteration sequence, which would also yield simple lower and upper bounds to the solution. Such bounds, due to the small value of a, are expected to be sufficiently sharp for all practical purposes, thus eliminating the need for costly

numerical integrations. This was, of course, achieved in the case of a single droplet. This phase of our work is well advanced, but the methods developed for handling the above equations are too involved to be discussed here with any detail.

II-6 Collection of drops - general case

The method of [39] has been recently generalized to account for the free boundary value problem for the three-dimensional heat equation, with any number of boundary conditions. It is thereby possible to reduce the general problem of a collection of drops (problem A) to that of solving a system of integro-differential equations; the number of these equations is equal to twice the number of drops.

In addition to terms defined in connection with the formulation of problem C, section II-1, we define:

$\underline{\xi}_n$ - position vector of point P_n (Fig. 3)

r_n, θ_n, ϕ_n - polar coordinates of a point with respect to a system of coordinates centered at P_n with cartesian axes obtained by translation of x, y, z axes.

$\underline{\omega}_n(\theta_n, \phi_n)$ - unit vector with origin at P_n

$d\omega_n$ - solid angle element on sphere centered at P_n

$\underline{\xi} = \underline{\xi}(\theta_n \phi_n, \tau) = \underline{\xi}_n + \underline{\omega}_n \rho_n(\tau)$, point on sphere $S_n(\tau)$ centered at P_n, of radius $\rho_n(\tau)$

$\underline{x}$ - position vector

$$u_n(t, \theta_n, \phi_n) = \frac{\partial}{\partial r_n} c \Big|_{\underline{x} = \underline{\xi}(\theta_n, \phi_n, t)} .$$

The equations for the radii of the drops are:

$$(1)\quad\begin{cases} \frac{1}{2}u_n(t,\theta_n,\phi_n) = \frac{\partial}{\partial r_n}\sum_m v_m \Big|_{\underline{x} = \underline{\xi}(\theta_n,\phi_n,t)} & \\ & n = 1,2,\ldots \\ \dot{\rho}_n(t) = \frac{a}{4\pi}\oiint u_n(t,\theta_n,\phi_n)\,d\omega_n & \\ \rho_n(0) = a_n \quad , & \end{cases}$$

where

$$(2)\quad\begin{cases} v_m = -\frac{1}{r_m}\sqrt{\frac{t}{\pi}}\left(e^{-\left(\frac{r_n-a_n}{2\sqrt{t}}\right)^2} - e^{-\left(\frac{r_n+a_n}{2\sqrt{t}}\right)^2}\right) \\ \qquad + \frac{1}{\sqrt{\pi}}\left(\int_{\frac{r_n-a_n}{2\sqrt{t}}}^{0} e^{-\sigma^2}d\sigma + \int_{\frac{a_n+r_n}{2\sqrt{t}}}^{-\infty} e^{-\sigma^2}d\sigma\right) \\ \qquad + \frac{1}{8\pi^{3/2}}\int_0^t \frac{\rho_n^2(t)}{(t-\tau)^{3/2}}\,d\tau \oiint u_n(\tau,\theta_n,\phi_n)e^{-\left(\frac{\underline{x}-\underline{\xi}}{2\sqrt{t-\tau}}\right)^2}\,d\omega_n \,. \end{cases}$$

Except for the special case of drops of initially equal radii, centered at the vertices of a cubic lattice, when equations (1) are the same for all n, we have here a large number, possibly infinite, of equations. To consider these equations with full rigor is a hopeless task, and an exact solution is not even desired considering that in study of aerosols initial data on individual drops cannot and need not be specified.

A first simplification of system (1) is obtained on the basis of a rather reasonable assumption that the distance d_{nm} between any two drops n and m is large compared to their radii. It then turns out that after integrating the first set of equations over the surface of the n'th drop, and dropping terms which can be neglected according to our assumption, $u_n(t,\theta_n,\phi_n)$ can be eliminated using the second set of equations. We get thus a set of equations of the form

$$(3)\quad \begin{cases} \dot{\rho}_n(t) = \dfrac{1}{\rho_n(t)}\left(G[\rho_n]-G^*[\rho_n]\right) + \sum\limits_m f[\rho_n(t),\rho_m(t),a_n,a_m,d_{nm}] & n=1,2\ldots \\ \rho_n(0) = a_n\ . \end{cases}$$

Here, $G[\rho_n]$ is a functional of ρ_n identical with the right hand side of eq. (3-1), whereas the other terms represent the corrections to eq. (3-1) due to the influence of other drops.

Equation (3) bears some similarity to equations obtained by Keller in trying to solve the problem of a collection of drops by a quasi-static method, and at the same time explains why the immodified quasi-static approach fails to lead to a coherent theory of evaporation, as noted in section II-5. As may be shown, the order of magnitude of $f[\rho_n,\rho_m,a_n,a_m,d_{nm}]$ for large d_{nm} is

$$f \sim d_{nm}^{-1}\, e^{-\frac{d_{nm}^2}{4t}}\ ,$$

hence, the series in (3) converge if t is finite. As we found already, the quasi-static approach is valid near evaporation time, hence for t large; see sections II-2 and II-3. In the process of carrying out the simplification, t is replaced by infinity in some places. If this is done, without further scrutiny in the series in eq, (3), the series becomes divergent, for its terms become asymptotic to d_{nm}^{-1} . This is the difficulty that was actually encountered by Keller.

If the drops are initially of the same radius, and their centers are at the vertices of a cubic latice, $\rho_n = \rho_m = \rho(t)$, any n and m, for all t. Eq. (3) reduce then to a single equation:

$$(4)\quad \begin{cases} \rho(t)\dot{\rho}(t) = G[\rho] - G^*[\rho] + \sum\limits_m f[\rho,\rho,1,1,d_{nm}]. \\ \rho(0) = 1\ . \end{cases}$$

This is the case considered already in section II-5, but the approximation is of a different nature, and should produce a more accurate result for ρ than eq. (5-2). Eq. (4) is of a form similar to those encountered already in preceding sections, and is now under investigation.

As to the general case, the system (3) is still too complicated for practical considerations. A further simplification can be obtained from the study of the equation determining the evaporation curve of a single drop, eq. (3-1). On this basis one can show that certain terms in (3) can be so simplified, without introducing an appreciable error in the final result, that the system (3) becomes a system of ordinary differential equations. This system is now being studied, and although we have no concrete results so far, it appears that the best approach would be that used in statistical mechanics, since the system studied is formally the same as the system of canonical equations of motion of large number of particles, with the $\rho_n(t)$ being interpreted as displacements. If this line of attack proves to be successful, the result will be in form giving the relative number of drops of radii $\rho(t)$ at time t, in terms of the initial density of radii. A result of this form is precisely what is desired in practical application.

We have, throughout this section, spoken exclusively of evaporation, but our considerations apply, with some modifications, to the case of condensation.

II-7. Effects of heat flow on process of evaporation[11].

In the above treatment of the evaporating drop (sec. II-2 and II-3) the effects of temperature have been neglected in order to simplify the discussion of the central mathematical difficulty encountered in finding the boundary of the evaporating drop. Now, however, using the techniques already developed, it is possible to attack the more general problem including the temperature effects.

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[11] by Ritger, P. D.

No final results have been obtained as yet, but we shall outline our approach and our plans for future work on this aspect of the problem.

In general, there is a decrease in temperature in the vicinity of the surface of an evaporating drop because of the absorption of the latent heat of vaporization. This results in a flow of heat in the interior of the drop and in the vapor region outside the drop. Moreover, the variation of temperature produces a change in the equilibrium pressure at the drop surface and hence affects the rate of evaporation.

Several possible formulations of this evaporation-temperature problem have been suggested, but the one that seems to lend itself most readily to treatment is one similar to that proposed by Kinzer and Gunn [26].

We consider an evaporating drop as in sec. II-1, but we now assume that the equilibrium concentration at the drop surface is a known function of the temperature. We further assume that the temperature $q(r,t)$ is initially constant everywhere, is continuous across the drop boundary, and satisfies the equation of heat conduction outside the drop, but is uniform (a function of t only) inside.

The mathematical formulation of this problem is as follows (in dimensionless coordinates introduced by transformations similar to those in II-1):

(1) $rc_t(r,t) = (rc(r,t))_{rr}$, $rq_t(r,t) = (rq(r,t))_{rr}$, for $r>\rho(t), t>0$.

(2) $c(r,0) = 0$, $q(r,0) = 0$

(3) $c(\rho(t),t) = F(q)$, for $0 \leq r \leq \rho(t)$, $t \geq 0$

(4) $c_r(\rho(t),t) = [1 + \frac{1}{\alpha} - F(q(\rho,t))]\dot{\rho}(t)$, $q_r(\rho(t),t) = -\delta\dot{\rho}(t)$, for $t>0$,

(5) $c(\infty,t) = 0$, $q(\infty, t) = 0$, $t \geq 0$,

(6) $\rho(0) = 1$, $F(0) = 1$,

where r is the radial distance from drop center, t the time, q the temperature, c the vapor concentration, F the known (empirical) function giving equilibrium concentration in terms of temperature, ρ the drop radius and α and δ positive constants. For convenience

we set the diffusion coefficient and the coefficient of thermal diffusivity numerically equal; an assumption of this type is unjustified and unnecessary for the continuation of our work and will be corrected in the final report on this piece of research.

This problem is attacked by the method of [39]. We obtain a pair of simultaneous integro-differential equations for $\rho(t)$ and $q(\rho(t),\ t)$; namely:

$$(7)\quad \begin{cases} \sqrt{\pi}\ \rho(t)\dot{\rho}(t) = G[\rho] + \sqrt{\pi}\ H[\rho,\ q] \\[2ex] q = K[\rho,\ q] \end{cases}$$

where

$$\left\{ \begin{aligned} & G[\rho] = \text{right hand side of equation (3-1).} \\ & H[\rho,q] = \alpha \left\{ (1-\rho\dot{\rho})(1-F(q)) + \frac{1}{\sqrt{\pi}} \int_0^t \left[\frac{(\rho(\sigma)[1-F])^{\cdot}}{\sqrt{t-\sigma}} + \right. \right. \\ & \qquad \left. \left. + \frac{[(1+\rho(\sigma)\dot{\rho}(\sigma)][1-F]z}{t-\sigma} \right] e^{-z^2} d\sigma \right\} \\ & K[\rho,\ q] = \delta\rho\dot{\rho} - \frac{1}{\sqrt{\pi}} \int_0^t \left[\frac{(\rho(\sigma)q)^{\cdot}}{\sqrt{t-\sigma}} + \frac{\delta\rho(\sigma)\dot{\rho}(\sigma)-q}{t-\sigma}\, z \right] e^{-z^2} d\sigma \\ & z = \frac{\rho(t)-\rho(\sigma)}{2\sqrt{t-\sigma}} \quad . \end{aligned} \right.$$

These equations are now being studied and we expect that a solution can be obtained by an extension of the iteration procedure which was used in solving eq. (3-1).

Assuming that we can obtain a solution to this problem, we should then be able to take account of temperature effects in the problem of the collection of drops (by means of the enclosed drop model, section II-5, or more accurately, as in section II-6). Also, we may be able to extend the method to include heat conduction in the interior of the drop or some of the other temperature effects that have been neglected.

General Remarks.

1. Personnel

The following are now actively participating in the research work on this project, either on part time or full time basis:

Dr. E. D. Corson,
Dr. B. Epstein,
Dr. I. I. Kolodner
Mr. M. Milgram
Mr. P. D. Ritger

It is planned to enlist the services of another research assistant within two months.

The work is coordinated by Dr. Kolodner, under the general supervision of Dr. R. Courant, Director of the Institute of Mathematical Sciences, N. Y. U. Other members of the Institute of Mathematical Sciences are frequently consulted. Dr. J. B. Keller, on leave of absence from New York University, at present head of the Mathematics Branch, Office of Naval Research, Washington D. C., who directed the research on this contract until September 1953, continues to take active interest in our work and serves in an advisory capacity. It is hoped that by the end of Summer 1954 he will rejoin this group as an active member.

Other members of the Institute who until recently devoted part of their time to this project are

Mr. M. Jordan
Dr. G. B. Whitham
Dr. B. Zumino

2. Attitudes

It is realized by the members of this group that the sponsoring agency is mainly interested in results in the form of workable formulae and discussions pertaining to the theoretical description of the Formation and Behavior of Aerosols. With this in mind, in the initial stage of our work we attacked various problems by approximation methods without paying too much attention to mathematical rigor. Correspondingly, results were often crude and not easily justifiable.

As it turns out, the problems encountered are of a complex mathematical nature. To treat them, various simplifying physical hypotheses are made, and then mathematical approximations follow. If the theoretical results thus obtained do not agree with experimental data, it is hard to determine whether the discrepancy is due to an oversimplified physical picture of the phenomena or to too crude mathematical treatment within the frame of the physical assumptions.

For this reason we attacked certain key problems with full mathematical rigor. This, of course, involved a great deal of work which at first may have seemed unwarranted in view of the practical objectives of this project. However, our efforts in this direction paid off handsomely. By obtaining a rigorous mathematical result, we are able to judge the validity of the physical model. Moreover, these considerations led to the discovery of powerful approximation methods which can be applied to the problems treated here and to many other problems of a similar nature.

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